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How gas separation membrane competes with chemical absorption in post-combustion capture

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Abstract

This paper describes an investigation for multi-stage systems used in coal-fired power plant. The whole work was divided into two steps: energetic and economic analyses. In the first step: on the basis of a serial concept, through varying the position of compressors and vacuum pumps, recycling the retentate of the 2nd membrane to the feed side of the 1st membrane, a cascade variant was developed and analysed. In the second step: an economic model was developed to calculate the capture cost of the cascade system. The total cost is composed of investment cost, operation and maintenance (O&M) cost and electricity cost. A correlation between the membrane parameters: selectivity & permeability and capture performance: energy consumption & capture cost was built up. Using Polyactive® membrane developed by GKSS with CO_2 permeance of 3 Nm^3/m^2hbar and CO_2/N_2 selectivity of 50, under the separation target of 70% degree of CO_2 separation and 95 mol% CO_2 purity, adopting the cascade membrane system in the 600 MW NRW-reference power plant, the specific energy consumption including CO_2 compression (110 bar, 30°C) is 256 kWh/$t_{separated\ CO2}$ with 6.4%-pts efficiency loss. The capture cost is 31 euro/$t_{separated\ CO2}$, which could be a promising solution as a retrofit for the existing power plants.

Keywords: post-combustion, gas separation membrane, multi-stage, energy consumption, economic analysis

1. Introduction

Although the chemical absorption method occupies a leading position in R&D on post-combustion with CCS [1], it has several inherent weaknesses: a) degradation of the solvent owing to the influence of the SO_2 and NO_x in flue gas and b) high energy consumption for the solvent regeneration process. As a technology competing with chemical absorption, the CO_2/N_2 gas separation membrane process for post-combustion capture is attracting more and more attention around the world. In comparison with the above mentioned weaknesses of chemical absorption, CO_2 gas separation membranes possess the following advantages: a) less environmental impact; b)

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can be designed as turnkey CO_2 separation equipment both for new-build power plants and as a retrofit for existing power plants. These are eminently important properties of the gas separation membrane process distinguishing it from the other post-combustion capture technologies.

Gas separation membranes used for post-combustion capture have been investigated by several groups independently [2-8]. In the present paper, multi-stage membrane processes are investigated in two steps: a) energy consumption and b) capture cost analyses. In the first step, by varying the position of compressors and vacuum pumps recycling the flue gas to the feed side, a cascade variant was developed and analyzed in detail. The cascade system was integrated with the 600 MW North Rhine-Westphalia reference power plant and compared with the chemical absorption process. In the second step, an economic analysis process was explored for a cascade membrane system for use in coal-fired power plant. A cost model was developed to make a further analysis of the cascade variant in view of the correlation between membrane parameters (selectivity, permeability) and system performance (energy consumption, capture cost).

In view of the R&D situation of the CO_2/N_2 gas separation membrane, the properties of Polyactive® polymer membranes with CO_2 permeance of 3 Nm^3/m^2hbar and CO_2/N_2 selectivity of 50 developed by GKSS, Germany [9], are used here. The PRO/II software (Simulation Science Inc.) was used for the simulation.

2. Investigation strategy

The investigation strategy is illustrated in Fig. 1. The whole simulation process is divided into two steps. In the first step, the influence of membrane parameters on membrane area to achieve the specified target is analyzed. This is combined with the evaluation of various process parameters, components and membrane arrangements. Membrane area and energy consumption are the two outputs of the analyses presented in this paper. In the second step, the effect of the variation of these parameters on the capture cost will be analyzed, especially the correlation between the membrane parameters (selectivity and permeability) and the capture performance (energy consumption and capture cost).

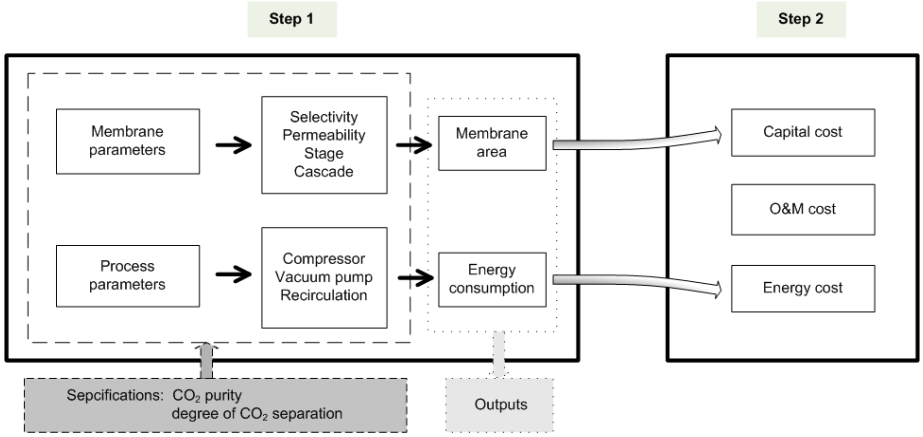

Fig. 1 Investigation strategy for multi-stage gas separation membrane systems

3. Reference power plant and simulation method

In the present work, a reference power plant termed the Reference Power Plant North Rhine-Westphalia (RKW-NRW) [10] was chosen for the analyses. The multi-stage polymer membranes should be installed after the SCR-DeNOx, dust removal (E-filter) and desulphurization (FGD) processes and prior to emissions passing through the cooling tower, analogous to amine stripping processes [11, 12].

The hard coal grade "Klein Kopje" was used to simulate the flow rate and the components of the flue gas for the multi-stage membrane calculation. The element analysis data of Klein Kopje coal are: C 65.5%, H 3.5 %, O 7.4%, N 1.5%, S 0.6%, ash 14.2%, moisture 7.3%; and the heat value is 25 MJ/kg. The coefficient of air excess (air-to-fuel ratio) was assumed to be 1.15. The basic data of RKW-NRW and the simulation results of the flue gas are listed in Table 1. The residue of the pollutant in the flue gas consists of approximately 50 vppm SO_2 and approximately 200 ppm NO_2.

The PRO/II (Simulation Science Inc.) software was used for the simulation. Different thermodynamic models for the energy balance calculation are available in PRO/II; for the case described here the Soave-Redlich-Kwong equation of state was chosen. The adiabatic efficiency of the compressors, expanders and vacuum pumps is assumed to be 85%. A detailed description of the membrane module in the PRO/II software was given in our previous paper [12]. A binary flue gas system – 14 mol% CO_2 and 86 mol% N_2 was simulated.

Table 1 RKW-NRW power plant basic data [10] and simulation results of the flue gas conditions after removal of the pollutants using Klein Kopje hard coal

Power plant RKW-NRW	
Output gross	600 MW
Output net	555 MW
Net efficiency	45.9 %
Steam parameters	285 bar/ 600°C / 620°C
Operation time	6000 h/year
Fuel input	1.0 Mt/year*
Investment costs	517.1 million euro
O & M costs	7.8 million euro/year
Fuel costs	41 euro/t
Electricity price	3.37 cent/kWh
Flue gas conditions after removal of the pollutants	
Pressure	1.05 bar
Temperature	50 °C
Flow rate	1.6 million m^3/h*
Main components	
CO_2	13.5 mol%*
N_2	70.1 mol%*
O_2	3.7 mol%*
H_2O	11.9 mol%*
Ar	0.8 mol%*

* simulated by PRO/II

4. Energetic analysis

4.1. Variations with compressor and vacuum pump

On the basis of the concepts of enricher and stripper, a matrix plan was developed for the probable arrangements of compressor and vacuum pump [13]. It is known that the enricher concept contributes to a higher CO_2 purity with a common separation degree than the stripper concept. In this paper, four variants V1-V4 (V1-V2: 1st membrane using compressor, V3-V4, 1st membrane using vacuum pump) of enricher are shown in Fig. 2. The relevant simulation results of the energy consumption and membrane area are listed in Table 2. In the simulation the flow rate of the feed gas is 100 Nm^3/h. The compressors are driven at 8 bar and the vacuum pumps at 30 mbar. The degree of CO_2 separation of each variant is defined as 70%, CO_2 purity as 90 mol%. Variant V3 provides a promising potential for fulfilling these requirements. This can be explained logically: using a vacuum pump for the 1st membrane to achieve a certain degree of CO_2 separation, then using a compressor for the 2nd membrane to obtain the desired CO_2 purity, this variant has an energy

advantage in comparison with those using a compressor for the feed flue gas, by means of which a considerable amount of energy is applied for N_2 compression.

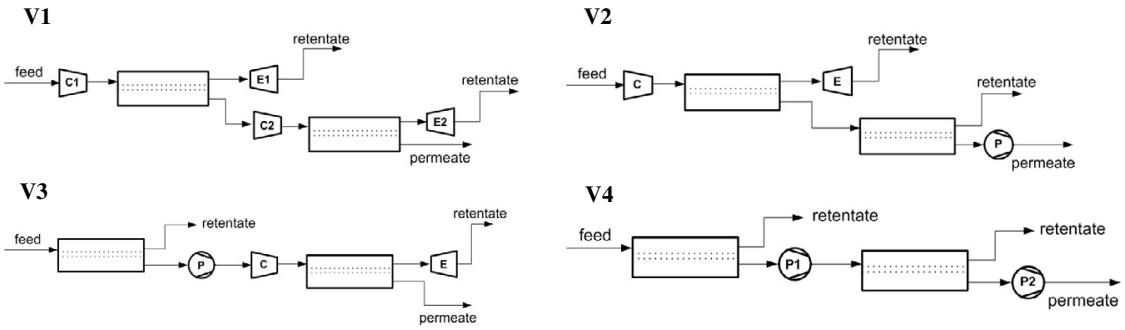

Fig. 2 Variations with compressor and vacuum pump for enricher concept

Table 3 Comparison of energy consumption of the 4 variants shown in Fig. 2, compressor: C = C1 = C2 = 8 bar, vacuum pump: 30 mbar

Variants	Separation degree [%]	CO_2 purity [mol%]	Area = (1^{st} + 2^{nd}) [m^2]	Specific energy [kWh/$t_{separated\ CO2}$]
V1	70	90	27	272
V2	70	90	42	296
V3	70	90	52	164
V4	70	90	71	204

4.2. Recirculation of flue gas

Another measure investigated here is recirculating the retentate of the 2^{nd} membrane back to the feed side of the 1^{st} membrane, on the basis of the simulation results of the single-stage membrane system [12], i.e. higher CO_2 composition in the feed gas enables higher CO_2 purity to be achieved after separation with the same membrane parameters and under the same operating conditions. Then a further retrofit V3-I was performed as shown in Fig. 3. Here the recirculation rate is defined as the ratio between the flow rate of the retentate of the 2^{nd} membrane and the total feed flow rate. By recycling the retentate of the 2^{nd} membrane to the feed side of the 1^{st} membrane, 95 mol% CO_2 purity can be reached. A detailed parametric study can be found in [13]. In order to explain how retentate recycling works within the V3-I cascade, an example is illustrated in Fig. 3, in which the flow rate and composition of different streams are labeled. Variant V3-I has most of the properties of the V3 cascade; a higher CO_2 purity can be achieved by recycling the flue gas within the system; it logically leads to higher energy consumption and a larger membrane area.

4.3. Comparison with MEA absorption

Variant V3-I was applied for 600 MW NRW-RKW including the CO_2 compression process (110 bar, 30°C). On the basis of the above simulation results, the following operating conditions were adopted: the vacuum pressure level of the 1^{st} membrane was kept 100 mbar, and the feed pressure of the 2^{nd} membrane remains at 4 bar. One aspect to be highlighted here is the question of the feasibility of a vacuum pump with a 30 mbar pressure level. Even large vacuum pumps will probably have a suction pressure of 50 mbar in the future, so the pressure drop within the module channels and connecting tubing should be considered additionally, which leads to reasonable vacuum pressure level of 100 mbar. The different degrees of CO_2 separation 50%, 70% and 90% are simulated with a uniform CO_2 purity of 95 mol%. The specific energy both for membrane capture and CO_2 compression process, so as to the efficiency loss are listed in Table 4.

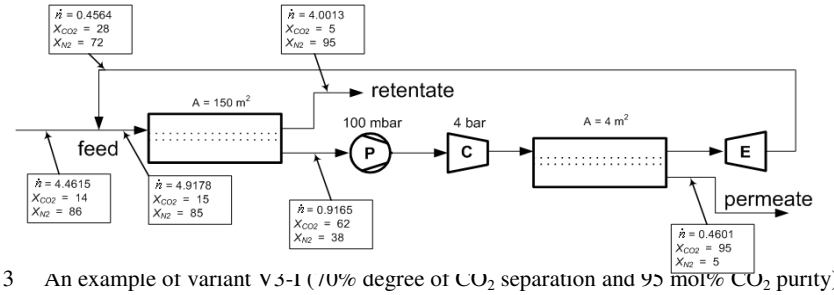

Fig. 3 An example of variant V3-I (70% degree of CO_2 separation and 95 mol% CO_2 purity), $\dot{n}_{feed} = 100 \quad Nm^3 \cdot h^{-1} = 4.4615 \quad kmol \cdot h^{-1}$

The energy penalty of the current MEA technologies ranges from about 8~14 percentage points for different types of power plants [14, 15]. Updated simulation results show that the capture process of MEA absorption consumes almost 10 percentage points of efficiency by adopting a similar power plant type [16, 17]. The efficiency losses of MEA absorption with 50%, 70% and 90% degrees of CO_2 separation including the CO_2 compression process (110 bar, 30°C) are listed in Table 5.

Table 4 Integration with NRW reference power plant using variant V3-I, separated CO_2 compressed to 110 bar, 30°C

Pressure		Separation degree [%]	CO_2 purity [mol%]	Membrane area × 10^6 [m^2]		Specific energy for capture [kWh/t_{CO2}]	Specific energy for compression [kWh/t_{CO2}]	Efficiency loss [%-pts.]
1st mbar	2nd bar			1st	2nd			
100	4	50	95	1.13	0.04	**124**	105	**4.1**
	4	70	95	2.39	0.06	**151**	105	**6.4**
	4	90	95	6.37	0.08	**244**	105	**11.4**

Table 5 Efficiency loss of MEA absorption with 50%, 70% and 90% degree of CO_2 separation, separated CO_2 compressed to 110 bar, 30°C [16, 17]

Separation degree [%]	CO_2 purity [mol%]	Specific energy for capture [kWh/t_{CO2}]	Specific energy for compression [kWh/t_{CO2}]	Efficiency loss [%-pts.]
50	99	220	100	**5.8**
70	99	220	100	**8.2**
90	99	220	100	**10.5**

It can be observed that the analyzed cases of variant V3-I have an energetic advantage in comparison with MEA absorption at 50% and 70% degree of CO_2 separation. This leads to a potential tendency that the gas separation membrane could be an important capture option as a retrofit for existing power plants, considering the above mentioned degrees of CO_2 separation.

5. Economic analysis

Applying a gas separation membrane system for post-combustion, the following cost factors should be considered: a.) capital cost (including membrane, frame, compression equipment and heat exchanger); b.) O&M cost; and c.) energy cost. An investigation of the literature [5, 18, 19] showed that the capture cost for MEA absorption is in the range of 30~50 euro/$t_{separated\ CO2}$.

Referring to work by a Dutch group [5, 6], in the present paper a similar simulation method was used to calculate the capture cost using the 600 MW NRW reference power plant. Table 6 lists 12 equations applied to determine the total capture cost and CO_2 specific separation cost. The relative cost and process parameters are shown in Table 7. Here the membrane cost is set to 50 euro/m^2 and the membrane frame, e.g. casing, valve, tubing, is calculated using equation (2) in Table 6. The nomenclature can be referred in the paper [13].

Table 6 Equations applied to determine specific CO_2 separation cost [5, 6]

Estimated investments *I* (components)		
$I_m = A \cdot K_m$	(1)	Membrane cost
$I_{mf} = (A/2000)^{0.7} \cdot K_{mf}$	(2)	Permanent membrane frame cost
$I_c = K_{c1} \cdot F_h + K_{c2} \cdot F_h$	(3)	Compressor cost
$I_{vp} = K_{vp} \cdot F_h$	(4)	Vacuum pump cost
$I_{ex} = P_{ex} \cdot K_{ex} \cdot F_h$	(5)	Expander cost
$I_{he} = C_{he}$	(6)	Heat exchangers and cooling facilities
Energy consumption of compression equipment *P*		
$P_{tot} = \sum P_c + \sum P_{vp} - \sum P_{ex}$	(7)	Total energy consumption
Annual costs C		
$C_{cap} = (\sum I_c + \sum I_{vp} + \sum I_{ex} + \sum I_{he} + I_{mf}) \cdot a + I_m \cdot a_m$	(8)	Capital cost
$C_{O\&M} = 0.036 \cdot (\sum I_c + \sum I_{vp} + \sum I_{ex} + \sum I_{he}) + 0.01 \cdot (I_m + I_{mf})$	(9)	O&M cost
$C_{en} = t_{op} \cdot P_{tot} \cdot K_{el}$	(10)	Energy cost per year
$C_{tot} = C_{cap} + C_{en} + C_{O\&M}$	(11)	Total cost
Specific CO_2 separation cost $\dot{C}_{CO_2}$		
$\dot{C}_{CO_2} = C_{tot} / M_{CO_2,ann,separated}$	(12)	

Table 7 Assumptions for cost and process parameters [5, 6, 10, 20]

Parameter	Value	Unit	Parameter	Value	Unit
K_m	50	euro/m^2	K_{mf}	0.25	million euro
K_{c1}	3	million euro	K_{ex}	0.3	euro/watt
K_{c2}	30	million euro	K_{vp}	$4K_{c1}$	million euro
C_{he}	3.5	million euro	F_h	1.8	-
a	0.064	-	a_m	0.225	-
t_{op}	6000	hour	K_{el}	3.37	cent/kWh

The depreciation time for the components of compressor, expander, vacuum pump, heat exchanger and membrane module is 25 years, and the lifetime of the membrane is 5 years; the O&M cost of the components of compressor, expander, vacuum pump and heat exchanger is assumed to be 3.6% of their capital cost, and for the membrane and membrane frame the O&M cost is taken as 1% of their capital cost. Here the compressor cost is composed of two parts: for capture and for CO_2 compression. The compressor for capture is related to the flue gas of 2~8 bar and a vacuum pump of 100 mbar. It is assumed that the vacuum pump costs 4 times as much as the compressor ($K_{vp} = 4K_{c1}$). The electricity price here is 3.37 cent/kWh. One aspect to be emphasized is that this price is the current power cost. The capture cost calculated here shows the CO_2 separation expense using the existing infrastructure. The capture cost was calculated for the variant V3-I, 70% degree of CO_2 separation, listed in Table 8. It is obvious that the capital cost, mainly from the membrane cost, dominates the total capture cost.

Fig. 4 shows the results of a parametric study of membrane selectivity, when the CO_2 permeance is defined as 3 and 5 $Nm^3m^{-2}h^{-1}bar^{-1}$. It can be observed that by increasing membrane CO_2/N_2 selectivity from 20 to 40, the energy consumption is effectively decreased, actually more than halved. However, from 40 to 80, this decreasing tendency is obviously slowed down: lines of open triangle for specific energy and open squares for capture cost. It should be mentioned here that with a CO_2/N_2 selectivity of 20, the recirculation rate is quite high at 30%, so that a large membrane area (1st membrane) is required to reach the required degree of CO_2 separation. When the selectivity is decreased to 40 and 60, the recirculation rate is accordingly reduced to 13.5% and 8.5%, respectively.

Table 8 Capture costs for the V3-I under the conditions: 1st membrane permeate pressure 100 mbar, 2nd membrane feed pressure 4 bar, 70% degree of CO_2 separation with 95 mol% CO_2 purity

Cost		Unit	Value
Specific CO_2 separation cost		euro/t_{CO2}/year	**31**
Total cost		million euro/year	**55.8**
Capital cost			35.3
	Membrane	million euro	122.2
	Membrane frame		36.2
	Compressor		59.4
	Vacuum pump		21.6
O&M cost		million euro/year	4.7
Energy cost			15.8

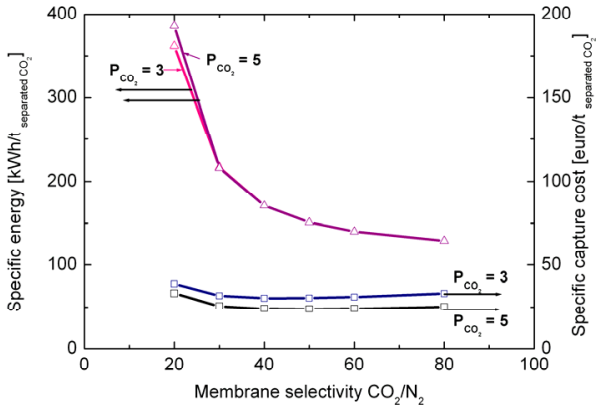

Fig. 4 Influence of membrane parameters (permeability & selectivity) on energy consumption, capture cost

From the simulation results, it is known that the Polyactive® membrane with a CO_2/N_2 selectivity of 50 and a CO_2 permeance of 3 $Nm^3m^{-2}h^{-1}bar^{-1}$ is attractive for a future gas separation membrane capture process. To realize the separation target of 95 mol% CO_2 purity and 70% degree of CO_2 separation, variant V3-I consumes 256 kWh/$t_{separated\ CO2}$ specific energy, with an efficiency loss of 6.4 percentage points and 31 euro/ $t_{separated\ CO2}$ capture cost for NRW-RKW.

6. Conclusions

Gas separation membrane capture used for post-combustion, as a competing technology, possesses the advantages of end-of-pipe application, and of less environmental impact than the chemical absorption method. The compact and modular structure makes it flexible in use and could be a promising option for a retrofit. The cascade concept developed in this paper is driven by electrical energy, which can be used as turnkey equipment for the application.

Process investigation provides us with the following knowledge of the system:

- A cascade arrangement makes it possible to reach high CO_2 purity;

- Owing to the feasibility of large-scale vacuum pumps and the reality of the pressure drop within the system, a 50% or 70% degree of CO_2 separation of the investigated cascade variant is attractive considering both energy consumption and capture cost;
- Membrane selectivity and permeability decide the CO_2 purity and the degree of CO_2 separation in a single-stage membrane, respectively, and strongly influence the energy consumption (electricity used to drive the compression machines) and total membrane area for a multi-stage membrane system, concerning the energy cost and capital cost, respectively. There is a trade-off balance between these pairwise parameters.

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References

[1] E. Blomen, C. Hendriks, F. Neele, Capture technologies: Improvements and Promising Developments, Energy Procedia, 1 (2009) 1505-1512.
[2] R. Bounaceur, N. Lape, R. Denis, C. Vallieres, E. Favre, Membrane processes of post-combustion carbon dioxide capture: A parametric study, Energy, 31 (2006) 2220-2234.
[3] E. Favre, Carbon dioxide recovery from post-combustion processes: Can gas permeation membranes compete with absorption?, J. Membr. Sci., 294 (2007) 50-59.
[4] M.-B. Hägg, A. Lindbråthen, CO_2 capture from natural gas fired power plants by using membrane technology, Ind. Eng. Chem. Res., 44 (2005) 7668-7675.
[5] C. Hendriks, Carbon dioxide removal from coal-fired power plants, Kluwer Academic Publishers, Dordrecht/Boston/London, 1994.
[6] J.P. van der Sluijs, C.A. Hendriks, K. Blok, Feasibility of polymer membranes for carbon dioxide recovery from flue gases, Energy Convers. Manage., 33 (1992) 429-436.
[7] T.C. Merkel, H. Lin, X. Wei, R. Baker, Power plant post-combustion carbon dioxide capture: An opportunity for membranes, J. Membr. Sci., doi:10.1016/j.memsci.2009.10.041 (2009).
[8] A. Brunetti, F. Scura, G. Barbieri, E. Drioli, Membrane technologies for CO_2 separation, J. Membr. Sci., doi:10.1016/j.memsci.2009.11.040 (2009).
[9] W. Yave, A. Car, S.S. Funari, S. Nunes, K.V. Peinemann, CO_2-Philic Polymer Membrane with Extremely High Separation Performance, Macromolecules, 43 (2010) 326–333.
[10] Konzeptstudie: Referenzkraftwerk Nordrhein-Westfalen (RKW NRW), VGB Power Tech e.V., Essen, Germany, February 2004.
[11] CO_2 capture ready plants, IEA Greenhouse Gas R&D Programme (IEA GHG), 2007/4, May 2007.
[12] L. Zhao, E. Riensche, R. Menzer, L. Blum, D. Stolten, A parametric study of CO_2/N_2 gas separation membrane processes for post-combustion capture, J. Membr. Sci., 325 (2008) 284-294.
[13] L. Zhao, E. Riensche, L. Blum, D. Stolten, Multi-stage gas separation membrane processes with post-combustion capture: energetic and economic analyses, J. Membr. Sci., 359 (2010) 160-172.
[14] B. Metz, O. Davidson, H. de Coninck, M. Loos, L. Meyer, IPCC Special Report on Carbon Dioxide Capture and Storage, Cambridge University Press, United Kingdom & New York, USA, available in full at www.ipcc.ch, 2005.
[15] H.J. Herzog, What future for carbon capture and sequestration?, Environ. Sci. Technol., 35 (2001) 148-153.
[16] P. Galindo-Cifre, K. Brechtel, S. Hoch, H. García, N. Asprion, H. Hasse, G. Scheffknecht, Integration of a chemical process model in a power plant modelling tool for the simulation of an amine based CO_2 scrubber, Fuel, 88 (2009) 2481-2488.
[17] W. Arlt, Thermodynamical optimization of solvents for CO_2 absorption, Workshop CO_2-Capture, -Utilization and –Sequestration, Status and Perspectives, PROCESSNET, DECHEMA-Haus, Frankfurt am Main, Germany, January 21st-22nd, 2008.
[18] D. Singh, E. Croiset, P.L. Douglas, M.A. Douglas, Techno-economic study of CO_2 capture from an existing coal-fired power plant: MEA scrubbing vs. O_2/CO_2 recycle combustion, Energy Convers. Manage., 44 (2003) 3073-3091.
[19] M.R.M. Abu-Zahra, J.P.M. Niederer, P.H.M. Feron, CO_2 capture from power plants Part II. A parametric study of the economical performance based on mono-ethanolamine, International Journal of Greenhouse Gas Control I(2007) 135-142.
[20] MEM-BRAIN Alliance (Gas separation membranes for zero-emission fossil power plants), 18 research institutions and 5 industrial partners Coordinator: IEF-1, Forschungs Zentrum Jülich GmbH, Oct. 2007 - Jun. 2011.